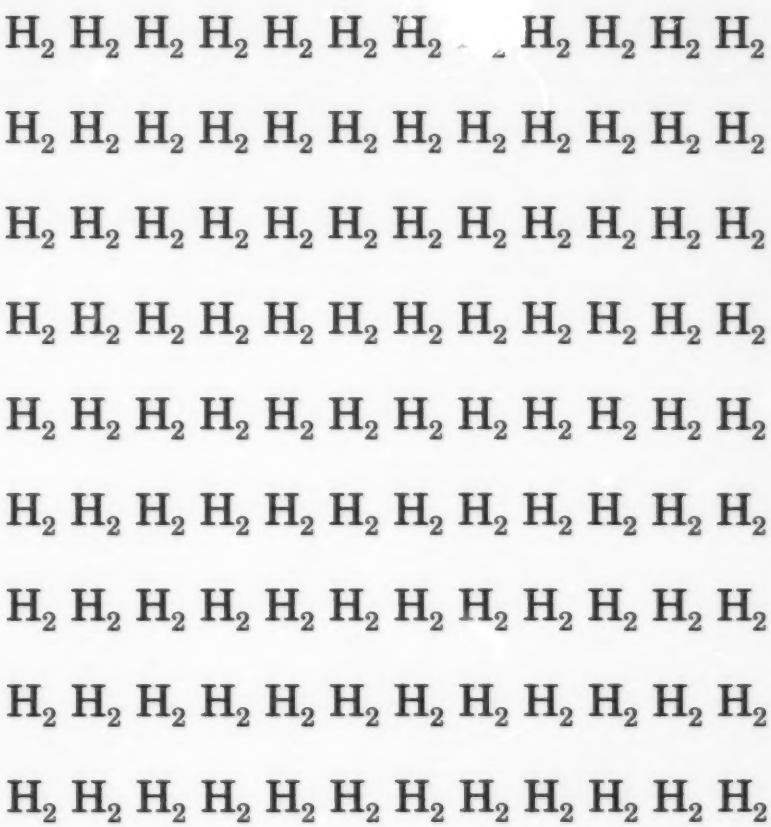
Alberta Hydrogen Research Program

Annual Review 1991/92



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Director's Report

More than half the hydrogen consumed in Alberta is used in the province's ammonia and methanol industries. The remainder is used in food processing, metallurgy, petrochemicals, and for upgrading heavy oil and bitumen. In particular, increasing amounts of hydrogen are being used in the heavy oil and bitumen sectors; this category now represents 22 per cent of the total provincial consumption. As well, recent growth in the size of Alberta's pulp and paper industry has been accompanied by a greater demand for hydrogen, which is used as a feedstock for hydrogen peroxide that is used to make paper. Since the products of all these industries are sold in world markets, their prices are under constant pressure from competitors. This means that manufacturers continually strive to lower their production costs and raise their production efficiencies. One way these twin goals can be achieved is to make one of the required feedstocks--hydrogen--available at lower prices, and improve how hydrogen is used in these various processes.

Currently, Alberta uses the cheapest method available for producing hydrogen--steam methane reforming. However, this process consumes a nonrenewable resource (natural gas) and produces carbon dioxide as a by-product. In addition to the environmental and resource factors, there is another concern: upgrading processes that are evolving for bitumen treatment and new refinery standards will require more hydrogen than is now used. This means that hydrogen production costs must come down. Taking all these issues into consideration, the Alberta Hydrogen Research Program aims to develop methods for producing hydrogen from hydrogen sulphide gas. The latter substance is widespread in Alberta, either present in many natural gas deposits or produced as a by-product of heavy oil and bitumen upgrading.

A commercial process for dissociating hydrogen sulphide into its two components--hydrogen and sulphur--will not only produce two valuable commodities from an otherwise hazardous chemical, but no other harmful environmental emissions are likely to be formed as by-products. Also, an effective process for dissociating hydrogen sulphide will have a direct effect on the processing of natural gas that contains significant amounts of hydrogen sulphide. For example, a resource such as the Bearberry sour gas field could produce enough hydrogen (along with sulphur) to feed a commercial-scale ammonia plant. This would diversify a resource that is totally dependent on sulphur markets.

The removal of sulphur (called hydrodesulphurization) from heavy oil and bitumen generates significant quantitites of hydrogen sulphide. Currently, this gas is converted to sulphur and water using the Modified Claus process, which is also used at natural gas plants scattered throughout Alberta. Based on the 580 000 tonnes of sulphur produced each year from Alberta's oil sands, recovering and recycling hydrogen from the hydrogen sulphide produced during hydrodesulphurization could produce an annual savings of up to $18 million.

Also, the standard bitumen upgrading process uses approximately 1 200 standard cubic feet (scf) of hydrogen for each barrel of bitumen. Upgrading converts bitumen, valued at approximately $8 a barrel, to an upgraded product that is sold for approximately $20 a barrel. Future high-conversion processes are expected to use approximately 2 200 scf of hydrogen per barrel of bitumen. Given the current low prices being paid for oil, less expensive hydrogen and more efficient use of it are needed to make synthetic crude oil competitive.

This then is the background for the Alberta Hydrogen Research Program. The focus is on making Alberta's energy industry more efficient, and using hydrogen as a principal link for integrating the oil and gas industry and diversifying Alberta's industrial base. It is appreciated that this approach differs from that used in hydrogen research activities in many parts of the world, where the emphasis is on hydrogen as a fuel and energy commodity. In Alberta's case, however, it is believed that all the components needed to produce, distribute and use hydrogen safely, more efficiently and at less cost by industry are the same elements needed to position Alberta at the forefront of the hydrogen-fuel industry.

The Alberta Hydrogen Research Program has now completed its third full year of operation, with a total of 20 research projects initiated. Thus far, the projects have all related directly to the production or use of hydrogen in the current petroleum and petrochemical industries of Alberta, and all were proposed and partly supported by these industries. Also, the contributions by industry to projects that are funded jointly with government rose in terms of overall percentage, reaching 50 per cent this year.

The program, however, is not limited to the oil and gas sector because it can embrace other industries that use hydrogen.

Perhaps the highlight of the year was the Hydrogen Research Program Workshop held in Calgary in November 1991. It was attended by a standing-room-only crowd, and the middle management of Alberta's oil and gas industry was well represented. This interest in a subject that has traditionally not been accepted as essential for the economic well-being of the industry was most gratifying, especially considering the current economic state of the industry.

Also, the contributions by industry to projects that are funded jointly with government rose in terms of overall percentage, reaching 50 per cent this year.

M.E. Torres
Director, Hydrogen Research
and Technology

Introduction

During 1987/88, a study to quantify current hydrogen production and consumption in Alberta revealed that Alberta industries produce 55 per cent and consume 63 per cent of all the hydrogen in Canada.

It was noted that most of the anticipated growth in hydrogen use is expected to arise from upgrading heavy oil and bitumen. Because the costs of hydrogen production currently represent a significant portion of overall upgrading costs, and the energy industry had previously expressed interest in working with government to lower these costs, a joint industry/government hydrogen technology research and development program was initiated in 1988/89. It was funded for three years on a cost-shared basis by the Alberta/Canada Energy Resources Research Fund (A/CERRF) and industry.

This was the final year of funding by A/CERRF, and the first year of a transition to government funding through the Alberta Department of Energy. The former name of the program--Hydrogen Technology Research Program--was changed to Alberta Hydrogen Research Program.

Currently, the program comprises the following components, each of which represents an aspect of hydrogen technology that requires commercial-scale development within 10 years.

The components are:

- advanced or alternative hydrogen production technologies;
- production technologies that work synergistically with those being developed in a separate coal research program;
- hydrogen separation technologies;
- large-volume hydrogen storage;
- transportation/distribution of hydrogen;
- optimum end-uses of hydrogen; and
- materials and safety technology.

From 1989/90 to 1990/91, 17 research projects were initiated and nine were completed. In 1991/92, three new projects were added to those that were continued from previous years. Including the hydrogen technology inventory study that was completed in 1988, 21 projects have been funded thus far.

The following section, Review of Program Projects--1991/92, provides detailed information about individual projects and their achievements. The financial aspects of past and present projects are found in Appendix B.

Review of Program Projects--1991/92

Hydrogen Production

Water (H_2O), methane (CH_4) and hydrogen sulphide (H_2S) are considered to be the three primary sources for hydrogen production in the immediate future in Alberta. This does not imply that all other potential sources are excluded, but rather that these three feedstocks represent the richest, cheapest and most readily available sources.

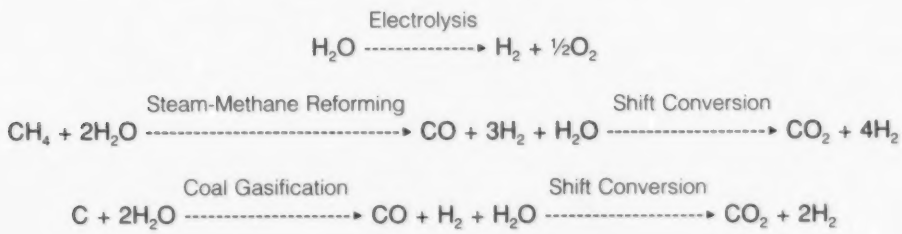
In western Canada, steam-methane reforming (SMR) is the most important and best developed hydrogen production method, but it is not necessarily the least expensive option for Alberta. It also generates carbon dioxide as a by-product, a fact that is of some concern to a world worried about atmospheric warming.

Nonetheless, whenever alternative hydrogen production technologies are proposed, they are compared to SMR. This requires evaluation of several factors, including process chemistry, energy input, process engineering, product separation and purification, and by-product uses.

In the following section on hydrogen production research, five projects are described. All represent attempts to develop new or improved hydrogen-producing systems that have commercial potential in the near future.

Thus far, all these investigations have focused on making hydrogen from hydrogen sulphide. The reasons for this direction are: the gas is abundant in Alberta; it must undergo some form of treatment before it is released to the environment; the current treatment method recovers only one of the two possible commodities; and hydrogen production from hydrogen sulphide does not release carbon dioxide. This latter characteristic, which is common to processes that electrolyse water to make hydrogen and oxygen, is often touted as the principal advantage of electrolysis over SMR.

CONVENTIONAL SOURCES OF HYDROGEN


Electrolysis
$H_2O \dashrightarrow H_2 + \frac{1}{2}O_2$
Steam-Methane Reforming
Shift Conversion
$CH_4 + 2H_2O \dashrightarrow CO + 3H_2 + H_2O \dashrightarrow CO_2 + 4H_2$
Coal Gasification
Shift Conversion
$C + 2H_2O \dashrightarrow CO + H_2 + H_2O \dashrightarrow CO_2 + 2H_2$

A Claus Plant Modification for Hydrogen Production[1]

ALBERTA SULPHUR RESEARCH LTD., CALGARY

Hydrogen sulphide gas is commonly found in Alberta's natural gas, and is also present in the "off gas" produced by upgraders at Alberta refineries. Because hydrogen sulphide can be toxic at low concentrations, it must be treated before it can be released into the atmosphere. This treatment occurs in plants using the Modified Claus Process. They convert H_2S to elemental sulphur by a sequence of combustion and catalytic steps. The primary product of this reaction, sulphur, is a valuable commodity. Currently, more than five million tonnes of sulphur are produced annually in Alberta using this technology. All the hydrogen present in H_2S, however, ends up in the other product of the Modified Claus Process--water.

Since Modified Claus plants are scattered throughout Alberta, Alberta Sulphur Research Ltd. proposed that they be used to produce sulphur and hydrogen instead of sulphur and water. In keeping with this, a project was initiated in 1989 to develop technology allowing the high temperatures available in a typical Claus furnace to be used for direct thermal decomposition of a portion of the hydrogen sulphide feed to Modified Claus plants. Not only would this provide a method for producing hydrogen as an additional commodity from the hydrogen sulphide stream, but a requirement of this approach is that existing capital equipment be used and current Claus plant economics not be affected.

During the first two years of the project, well over 100 laboratory experiments were conducted to determine the effect of various reaction conditions on thermal cracking efficiency. Thus far, hydrogen yields of up to 25 per cent have been achieved in single passes at 900°-1 200°C, with residence times of one second or less. These temperatures are consistent with normal Claus operations. The studies also showed that catalysts can be effective in improving yields at lower temperatures.

Because some carbon dioxide and other gases are likely to accompany hydrogen sulphide in feed gas mixtures, thermal decomposition studies were conducted on CO_2-containing gas mixtures in the presence or absence of catalysts. The results indicated that these other components have relatively little influence on hydrogen yields, provided the gas contains more than 70 per cent H_2S.

Computer modelling of the effect of incorporating a thermal cracking system within a Claus facility indicated that little or no reduction in the overall sulphur-recovery efficiency of the Claus plant will occur. Indeed, certain aspects of the thermal cracking operation can have a beneficial effect on the Claus operation.

Design drawings have been prepared of a thermal cracking test unit that would be installed in an existing Claus plant. It was decided the device would be constructed from a high Co-Cr-Ni alloy known as Haynes HR-160. This alloy has been tested successfully to 900°C under conditions similar to those that will be encountered in a Claus furnace, but testing has not been done at 1 200°C.

The investigation has further shown that it is essential to develop a separation technique to recover hydrogen at or near the cracking temperature. This must be done to prevent back reaction from occurring that will convert the products back to H_2S. Thus, work started last year on a high-temperature ceramic membrane system. At year-end, some encouraging results had been obtained from microporous alumina ceramic membranes that had been impregnated with sodium silicate.

[1]The project received financial support from Alberta Sulphur Research Ltd. and A/CERRF.

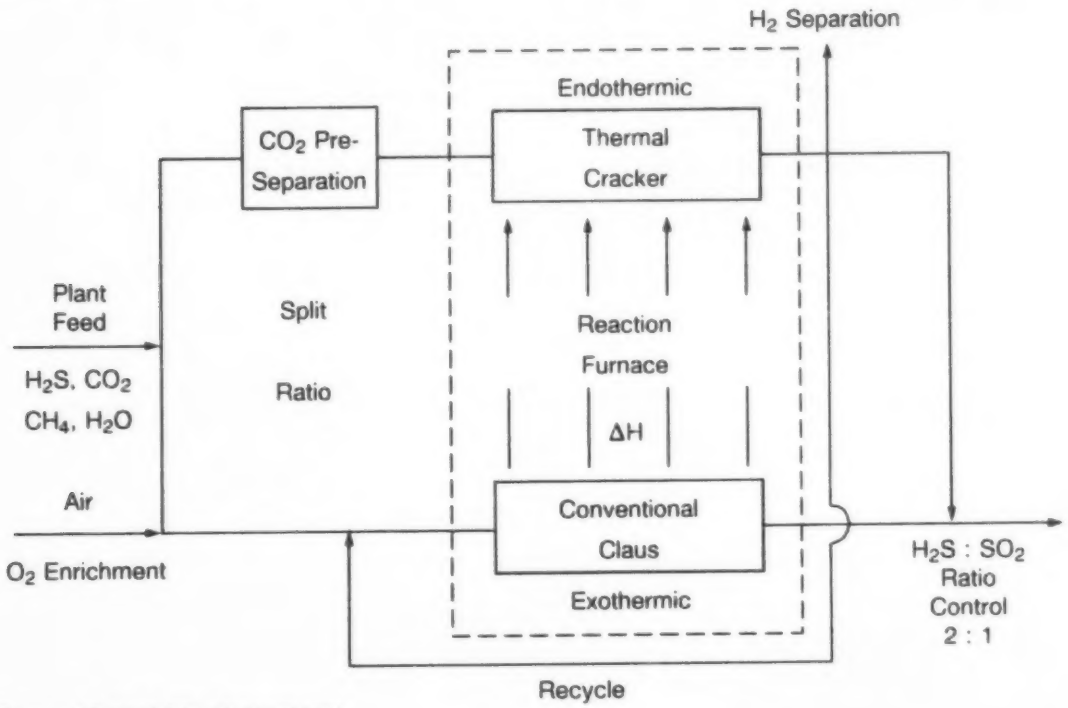
This year the work was continued by using a palladium membrane to separate mixtures of hydrogen and argon at temperatures ranging from 200° to 800°C. The separated gas was pure hydrogen and the measured flow rate through the membrane was similar to the fastest flow rate achieved for the best of the alumina membranes.

Despite these results, it was believed that the cost of a commercial-scale palladium membrane would be prohibitive. Therefore, a more promising direction was proposed for the work--that of using a composite membrane comprising a thin palladium film bound to a mechanically stronger porous ceramic.

A microporous alumina membrane was modified by *in situ* vapour phase hydrolysis of silicon tetrafluoride within the pore structure of the membrane. While passage of a 50:50 mixture of hydrogen and argon through the modified membrane resulted in a significant enrichment of the hydrogen fraction in the product mixture (74:26 H_2:Ar), the membrane was found to be unstable at the desired operating temperature of 1 200°C.

Nonetheless, another series of experiments in which preformed silica microspheres occupied the pore spaces in alumina membranes produced a significant enrichment of hydrogen, and operating these membranes at 900°-1 200°C did not adversely affect their stability. At year-end, a different type of alumina membrane was being prepared for testing.

Computer Simulation Model of a Modified Claus Front-End Furnace



(Source: Alberta Sulphur Research Ltd.)

Chemically Modified Electrodes for Hydrogen Sulphide Electrolysis[1]

ALBERTA RESEARCH COUNCIL, EDMONTON

The direct electrolysis of hydrogen sulphide is regarded by some researchers as the best potential method for producing hydrogen and sulphur on a commercial scale. However, sulphur deposition on the anode continues to be a problem. While some researchers claim to have overcome "anode blocking," they did it using electrical currents that are far too low for industrial requirements.

In a project initiated at the Alberta Research Council, the objective was to gain fundamental knowledge about the coproduction of hydrogen and sulphur by electrolysing aqueous solutions of hydrogen sulphide.

It has long been recognized that hydrogen sulphide can be scrubbed effectively in alkaline solutions, but previous attempts to achieve a succesful electrolysis have resulted in anode passivation caused by a blocking layer of sulphur. In this project, surface-modified electrodes were developed that will enable rapid transfer of electrons, while preventing adherence of sulphur. After proving the concept of chemically modified electrodes last year, development was extended toward finding superior zeolite-incorporated catalysts.

Several carbon electrodes modified with either metal-exchanged zeolites or transition metal complexes trapped inside the zeolites were studied. Current densities obtainable with some of these electrodes (e.g., NiY-carbon) are up to 150 mA/cm^2, which makes them suitable candidates for scale-up development.

The experiments in acidic solutions showed current densities that were approximately one-tenth those in alkaline solutions. This implies that future research should be directed toward alkaline solutions.

A brief, limited study of gas-phase electrolysis on partly immersed electrodes showed very slow reaction rates. These results are discouraging, but do not completely exclude the feasibility of the gas-phase reaction with different catalysts and under different conditions.

The effectiveness of the process was affected by a parasitic reaction on the cathodic side. While anodic materials have received most of the attention of researchers in this field, this work shows that the nature of the cathode is also important. It is recommended that in any future research involving H_2S electrolysis in alkaline solutions, the development of suitable cathodes be assigned a high priority.

This research project also proved that the performance of chemically modified anodes for H_2S electrolysis was not affected by the presence of other sour gas components, such as carbon dioxide and methane. Sulphur and electrolyte recovery remain as engineering considerations.

Based on these results, it was concluded that further development of a scaled-up process is warranted. The issue of cathode material and the overall current efficiency must be resolved, however. It is also recommended that sulphur recovery and electrolyte regeneration be discussed in the engineering context at a specific site or plant, rather than in general terms.

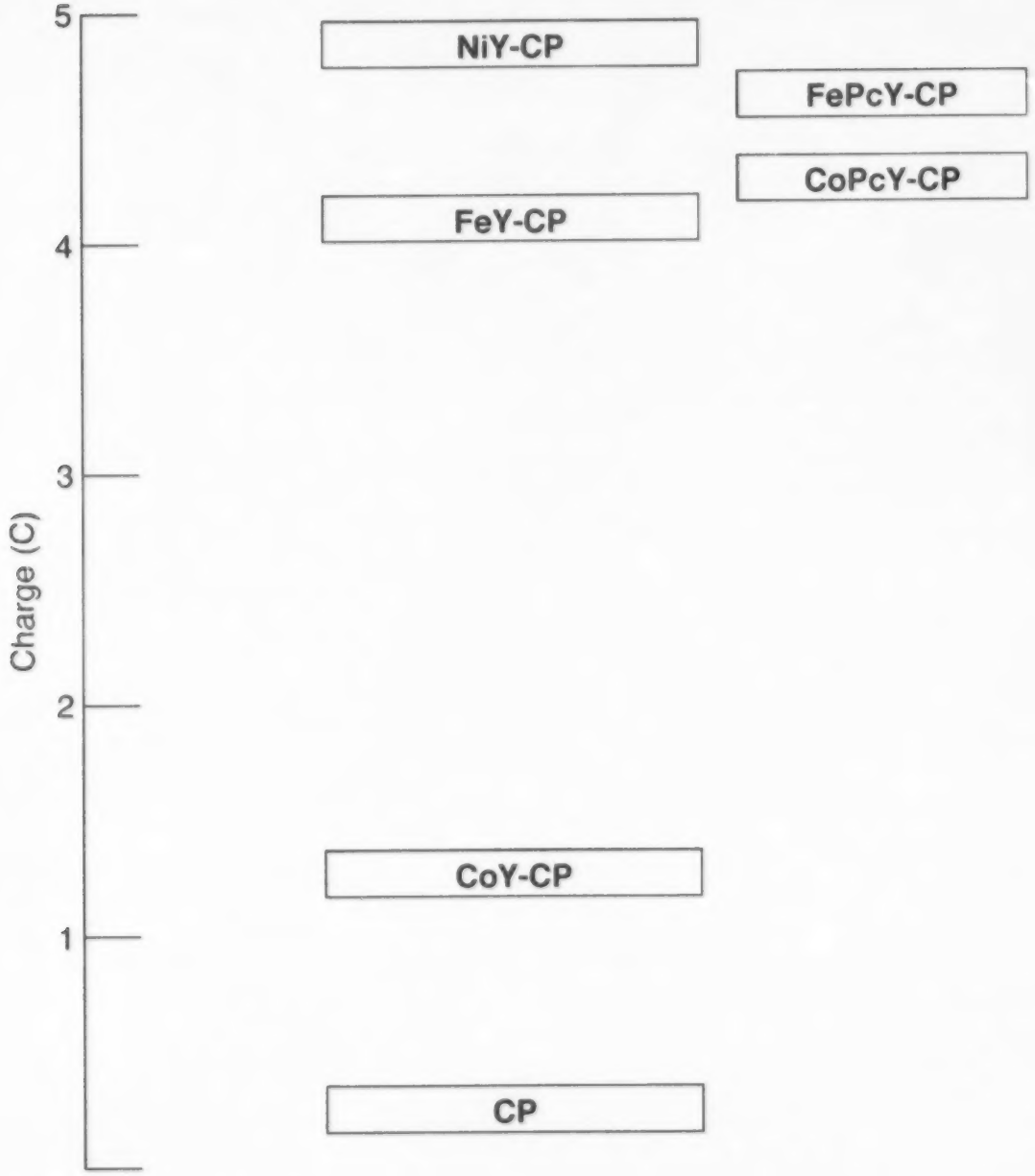
Publication

Petrovic, S. 1991. Chemically Modified Electrodes for the Electrolytic Production of Hydrogen and Sulphur from Hydrogen Sulphide. Alberta Research Council.

[1]The project was funded jointly by Alberta Research Council and Alberta Department of Energy.

Electrolysis Data Chart

Electrodes or Electrode Groups: Metal and Complex Exchanged Zeolites
Experimental Conditions: NaOH/NaHS(1M), +600mV, 600s



(Source: Alberta Research Council.)

The Carbon Monoxide Catalyzed Conversion of Hydrogen Sulphide to Hydrogen and Sulphur[1]

UNIVERSITY OF ALBERTA, EDMONTON

An alternative to electrolysis or thermal conversion of hydrogen sulphide to obtain hydrogen was explored in this project. The objective was to produce hydrogen from a two-step reaction between hydrogen sulphide and carbon monoxide, and then regenerate carbon monoxide for reuse.

Using a catalyst, one atmosphere pressure and a reaction temperature of 263°C, conversions to hydrogen and carbonyl sulphide, which represented up to 94 per cent of the thermodynamic equilibrium value, were achieved according to the following reaction:

$$H_2S + CO \rightleftharpoons H_2 + COS$$

In less than one minute residence time, 24.4 per cent of the H_2S was converted to H_2 and COS.

A second reaction step was proposed to convert carbonyl sulphide to elementary sulphur, as follows:

$$COS \rightleftharpoons CO + S$$

This reaction, however, has been found to be accompanied by a competing reaction, namely:

$$2\ COS \rightleftharpoons CO_2 + CS_2$$

Also, if any residual H_2S happens to be present, other competing reactions can occur:

$$CO_2 + H_2S \rightleftharpoons COS + H_2O$$

$$COS + H_2S \rightleftharpoons CS_2 + H_2O$$

During this investigation, a process was developed that could achieve a high selectivity for the desired production of elementary sulphur and carbon monoxide. At a reaction temperature of 750°C, product yields of 54 per cent were achieved. Details of the reaction conditions are not available, as patent protection is being sought.

At year-end, the reaction conditions were being optimized for industrial applications.

Publications

Strausz, O.P. 1992. The CO-Catalyzed Conversion of H_2S to H_2 and S. April 1, 1991 - March 31, 1992. University of Alberta.

Strausz, O.P. 1991. The CO-Catalyzed Conversion of H_2S to H_2 and S. University of Alberta.

[1]Funding during the first year of this project was provided by Gulf Canada Resources Limited and A/CERRF. During the second year, substantial funding was provided by the National Science and Engineering Research Council, in addition to Gulf Canada Resources Limited and the Alberta Department of Energy.

Cold Plasma Decomposition of Hydrogen Sulphide[1]

SHELL CANADA LIMITED (CALGARY) AND
ATOMIC ENERGY OF CANADA LIMITED
RESEARCH COMPANY (CHALK RIVER)

In this project, conducted by Atomic Energy of Canada Limited (AECL) in collaboration with Shell Canada Limited, microwave energy was used to initiate and maintain a hydrogen sulphide plasma. In this state, hydrogen sulphide can be decomposed into its elemental components, hydrogen and sulphur.

In studies reported in the scientific literature, successful decomposition of hydrogen sulphide has been proven, but at conditions that would not be practical for large-scale, commercial operations. Thus, one objective of the AECL project was to investigate the microwave-induced decomposition of hydrogen sulphide at conditions that are more commercially suitable, i.e., at higher pressure and with impure feeds.

During the first year of this project, a special apparatus was designed and constructed. It included a system for efficient removal of sulphur formed during the decomposition reaction, and a method for recirculating any unreacted hydrogen sulphide.

Hydrogen sulphide was converted to hydrogen and sulphur at various flow rates. At low gas flow rates, the decomposition yield was as high as 85 per cent. Also, effects of methane and carbon dioxide feed gas contaminants were measured on the percentage decomposition, product formation and the electric field at the plasma. The percentage decomposition of H_2S was maintained in the presence of carbon dioxide and increased in the presence of methane.

A lower frequency radiation source, developed during the first year of the project, was used during the second year to generate the plasma. This device allowed the formation of a cylindrically symmetrical hydrogen sulphide plasma that could be maintained to atmospheric pressure, and the percentage decomposition to hydrogen and sulphur was several points higher than was achieved last year with the higher frequency source. The inclusion of CO_2 and CH_4 as feed gas contaminants had little effect on the decomposition yield.

Some experiments were carried out on methods for quenching the decomposition reaction to minimize the recombination of hydrogen and sulphur to form H_2S. Also, a method for inserting probes into the plasma was developed to ascertain its temperature. This process allowed temperature and species profiles to be established, an important step in determining the characteristics of plasmas.

The work this year led to the conclusion that the lower frequency radiation source is suitable for operating at a commercial scale.

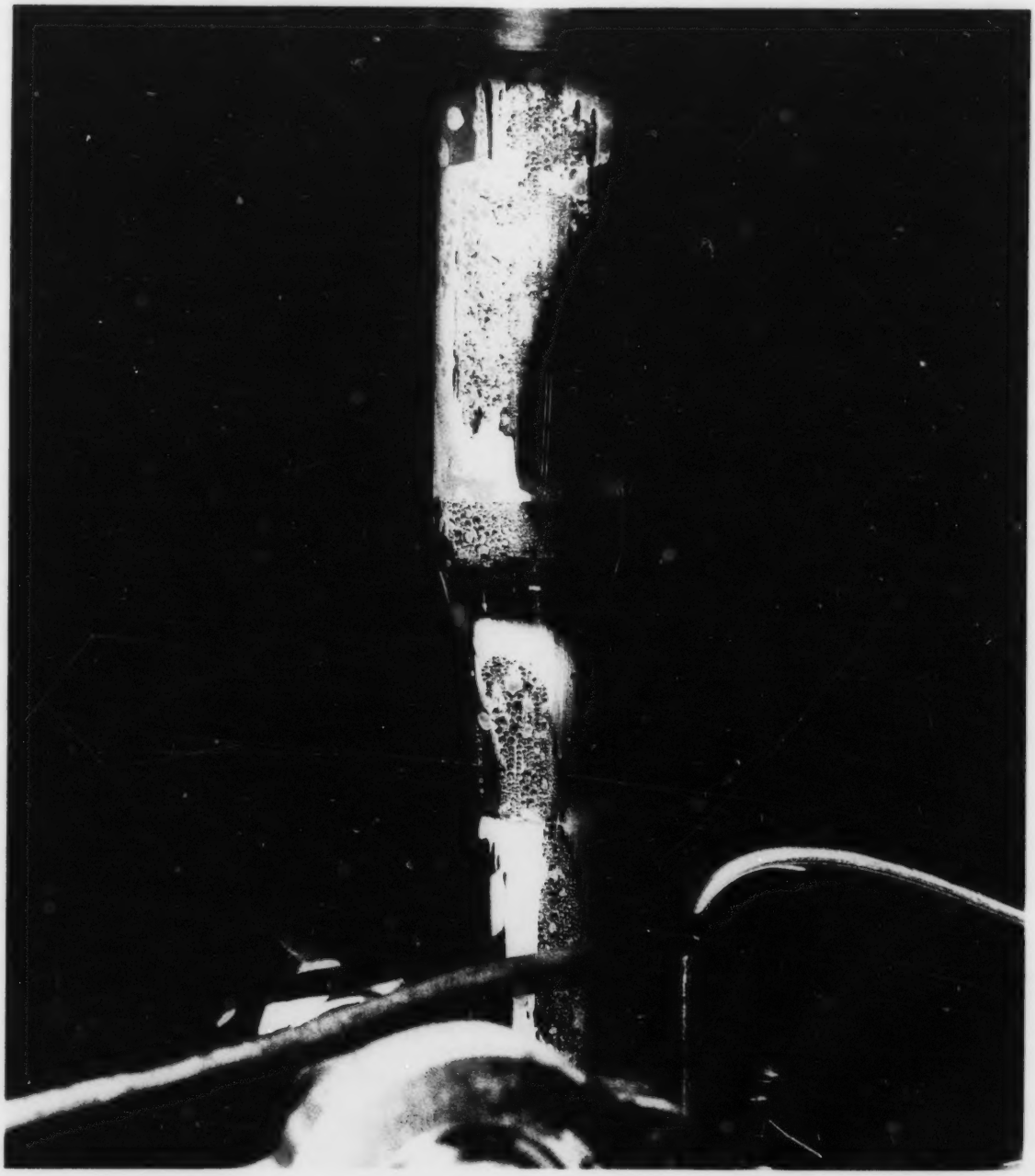
Next year, the economics of a commercial operation will be determined.

Publications

Craw, M.T., L.J. Cornett, K.D. McCrimmon and R.M. Hutcheon. 1991. Cold Plasma Decomposition of Hydrogen Sulphide. AECL Research/Shell Canada Limited.

Craw, M.T., L.J. Cornett, K.D. McCrimmon, R.P. Tremblay, J.H. Rowat and R.M. Hutcheon. 1992. Cold Plasma Decomposition of Hydrogen Sulphide. Annual Report 1991-92. AECL Research/Shell Canada Limited.

[1]The project was funded by Shell Canada Limited, Atomic Energy of Canada Limited, A/CERRF and the Alberta Department of Energy.



When a hydrogen sulphide plasma decomposed, one of the products--sulphur--deposited on the walls of the reactor. (Photo courtesy of AECL Research).

Microwave-Initiated Decomposition of Hydrogen Sulphide[1]

UNIVERSITY OF ALBERTA, EDMONTON

Microwave-initiated catalysis is a new process in which a metal-based catalyst is heated by microwave energy to a high temperature in the presence of one or more reactants. When a reactant contacts the surface of the catalyst, the reactant decomposes and then rapidly desorbs from the surface. The potential for the industrial conversion of methane to hydrogen has already been demonstrated using the process.

The objective of this project, which began in mid-1991, is to use the same approach for decomposing H_2S.

Thus far, a stream of pure H_2S has been decomposed in the presence of a metal-containing catalyst on an alumina support.

The two principal reactions are:

$H_2S \rightarrow H_2 + S$ (1)

and

$H_2S + M \text{ catalyst} \rightarrow MS + H_2$ (2)

Reaction (2) was shown to occur readily, but reaction (1), which is the desired reaction, was of only minor significance. Water also formed, indicating the desorption of water from certain components of the catalyst, or the reaction of the metal oxide in the catalyst with H_2S. Thus, at year-end other catalysts were being tested.

[1]Funding was provided by Canadian Occidental Petroleum Ltd. and the Alberta Department of Energy.

Publication

Strausz, O.P. 1992. Microwave-Initiated Decomposition of Hydrogen Sulfide. June 1, 1991 - March 31, 1992. University of Alberta.

Hydrogen Separation

Separation of produced hydrogen from unreacted feedstock or reaction by-products is one aspect of the Alberta Hydrogen Research Program that deserves special attention.

Although several straightforward hydrogen separation methods exist, using these technologies in combination with thermally or chemically energized hydrogen production processes is often inefficient. This inefficiency is frequently caused by large changes in the process temperature and pressure.

Also, some existing hydrogen-producing processes might become economically attractive if they could be combined with suitable separation methods.

For these reasons, two hydrogen separation projects have been initiated. One was completed last year and the other is described in the following section.

High-Temperature Separation of Hydrogen Using Novel Ceramic Membranes[1]

ALBERTA RESEARCH COUNCIL, EDMONTON

Thin-film polymeric materials are used commercially to separate the components of various gas mixtures. However, these separation media are not well suited for the high-temperature applications that are likely to be used in any new or improved hydrogen-production process in Alberta. A possible alternative is ceramic membranes based on porous metal oxides, such as alumina. Compared with polymeric membranes, they have a higher thermal stability (1 000°C versus <200°C), higher operating pressure, longer life and higher permeability. The principal disadvantage of ceramic membranes is their cost; approximately 10 times that of polymeric membranes.

Thus, a research project to develop novel ceramic membranes for high temperature separation of hydrogen was initiated at the Alberta Research Council in 1990. The principal objectives are to optimize preparation techniques for membranes based on zeolite/sol and alumina sol-gel, and to obtain performance data for tubes coated with these membranes. Included in this work is the preparation and characterization of Ti- and Zr-based membranes.

The membrane techniques developed in the project can result in membranes with an average pore size of <2 nm. This should provide considerable selectivity for separating hydrogen from other substances.

In preparing ceramic membranes, the two most important characteristics desired in the finished product are pore size and pore size distribution. Membranes must be free of cracks and thermally stable to achieve optimum separation. In previous investigations reported in the scientific literature, two preparation techniques have emerged: slip casting and sol-gel.

While the slip-casting technique is particularly suitable for supporting substances of various shapes, the pore size diameter of the resulting membrane is usually larger than 50 nm. This is regarded as too large for the kinds of mechanisms that are most desired for hydrogen separation.

In sol-gel techniques, a colloidal solution (or sol) is produced and applied to the support. Water is allowed to evaporate, forming a gel that is thermally treated to produce the desired film. Experiments have shown that this process is somewhat promising, based on the separation factors achieved thus far.

The most promising approach found in this study involved the use of zeolites. Not only could zeolite layers be bonded to ceramic supports, but the resulting membranes can provide separation through molecular sieving mechanisms.

The membranes were characterized by various techniques to determine their physical properties and performance. These tests included scanning electron microscopy, chemical composition analysis and gas permeability measurements. The slurries and sols from which the membranes were prepared were also analysed.

Several recipes for zeolite-based membranes with different concentrations of zeolite on the surface were investigated. The results showed that the maximum separation factor obtained for H_2/CO_2 using a zeolite-coated membrane was 2.43. Subsequent testing showed that zeolite membranes are stable at temperatures up to 750°C, and can be applied to larger tubes.

[1]Funded by Shell Canada Limited, Alberta Research Council and the Alberta Department of Energy.

The performance of tubes coated with alumina sol-gel was somewhat better than that of the commercially available ASTD5 tube, but inferior to that of zeolite-coated tubes. Further, it was not possible to apply the recipe to the surfaces of tubes, and the membranes were not sufficiently stable.

Ti and Zr sols were prepared using the corresponding alkoxides as the starting materials. Generally, the Ti-based membranes were more thermally stable than alumina-based membranes. Consequently, additional investigations of Ti-based composite membranes will be among the tasks to be carried out next year.

Publication

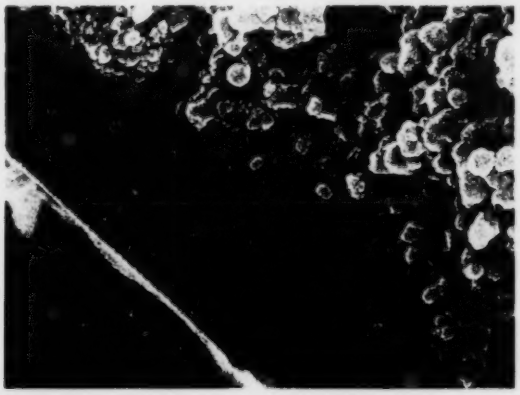
Rangwala, H.A. and L. Johanson. 1992. High Temperature Separation of Hydrogen Using Novel Ceramic Membranes. Alberta Research Council.

Smith, K.J and L. Johanson. 1991a. Development of Ceramic Membranes for High Temperature Separation of Hydrogen. A Review. Alberta Research Council.

Smith, K.J. and L. Johanson. 1991b. High Temperature Separation of Hydrogen Using Novel Ceramic Membranes. Alberta Research Council.

Cross-sections of membranes

Zeolite



Alumina

(Source: Rangwala, H.A. and L. Johanson. 1992. High Temperature Separation of Hydrogen Using Novel Ceramic Membranes. Alberta Research Council.)

Infrastructure and Storage/Transportation

The net hydrogen surplus currently existing at some Alberta facilities is expected to become larger as new or improved hydrogen production technologies are introduced. Fortunately, this surplus is expected to be offset by expansion of bitumen upgrading, increased synthetic fuel production and growth in the production of some specialty chemicals. All of these will consume hydrogen.

Thus, hydrogen needs to be moved from locations where its production exceeds on-site demand to places where it is needed, but not produced. This would involve some method for integrating producers and consumers.

Before an integrated producer/consumer network can be established, however, numerous technical issues must be addressed. One of the first requirements is to develop a strategy to link producers with consumers, including a staged development scheme.

Of equal importance is the need to investigate several storage and transportation aspects. These will depend on the physical state of hydrogen when it is stored or transported, and must recognize that hydrogen could be handled as a gas or a liquid.

Transportation of hydrogen could involve some combinations of truck, rail and pipeline. Like storage, this would require investigation of certain safety aspects and the physical effects of hydrogen on the storage or transportation medium.

During 1989/90, two studies related to these issues were undertaken and completed. In one, a computerized method for integrating hydrogen producers and consumers was developed at the Alberta Research Council. In the other study, a preliminary engineering investigation was made of the concept of storing hydrogen in salt caverns. Both studies produced positive and encouraging results and paved the way for one additional study that was carried out from 1990 to 1992. This latest investigation is described in the following section.

Hydrogen Transportation Using Present Pipeline Networks[1]

HYDROGEN INDUSTRY COUNCIL, CALGARY

An important component of Alberta's hydrogen research program is having the ability to transport hydrogen to markets in an economically and technically sound manner. As the demand for hydrogen grows, this need will become more urgent.

It is believed that hydrogen can be transported along with natural gas in the existing natural gas pipeline network that extends throughout much of Canada and the United States. Studies carried out thus far have indicated that transportation of gas/hydrogen mixtures is feasible, and only minor adjustments to mechanical systems are required to accommodate hydrogen.

In this project, led by the Hydrogen Industry Council and funded by several companies, A/CERRF and the Department of Energy, the objective was to investigate the market potential for hydrogen/natural gas mixtures that are transported in existing pipelines. The economics of the concept were examined in terms of hydrogen sources and uses. As well, a number of technical issues were addressed, including any adverse effects of gas mixtures on the mechanical integrity of pipelines and pumping equipment.

Individual contractors were hired to investigate the following components:

- thermodynamic and combustion properties of hydrogen and hydrogen/natural gas mixtures;
- purification, pipelining, storage and compression of hydrogen gas;
- fuel cell applications; and
- collection, distribution and uses for hydrogen in Alberta and beyond.

The investigation found that approximately 227 million standard cubic feet per day (scfd) of by-product hydrogen are burned as fuel at chemical plants in Alberta. This is hydrogen that might be used for higher value-added purposes if it were possible to transport it to locations where it is needed. Furthermore, indications suggest that this volume will increase to 354 million scfd by the year 2000.

There are two options for using this surplus hydrogen: use it in Alberta or sell it outside Alberta. In either option, a method for moving the hydrogen by pipeline is necessary. This could involve blending hydrogen with natural gas and transporting the blend in existing pipelines, or constructing a new pipeline to transport hydrogen that is 99.8 per cent pure at least. The latter option would be needed for California customers and the proposed HYPER pipeline linking Edmonton and Redwater.

For markets within Alberta, an investigation by Colt Engineering Corporation proposed that a collection system must be capable of collecting surplus hydrogen from plants as far away as Medicine Hat, Joffre and the Edmonton/Fort Saskatchewan area. Also, salt cavern storage near the Shell Canada refinery at Scotford should be included in the system. Furthermore, excess hydrogen-producing capacity exists at other Alberta plants that have shut down some production units as the result of the current economic climate. If these shutdown units were reactivated, they could help overcome any hydrogen shortages caused by temporary "outages" at the major sources of surplus hydrogen.

[1]Funding was provided by Gulf Canada Resources Limited, Northwestern Utilities Limited, Union Carbide Canada Limited, OSLO Alberta Limited (as agent for the OSLO Participants, namely Petro-Canada Inc., Alberta Oil Sands Equity, Pan Canadian Petroleum Limited, Esso Resources Canada Limited, Canadian Occidental Petroleum Ltd. and Gulf Canada Limited), Hydrogen Industry Council (project operator), A/CERRF and the Alberta Department of Energy.

Thus, the supply situation in Alberta could accommodate any expanded use of hydrogen in the province. The creation of a hydrogen-supply grid was recommended whenever a major new demand for hydrogen arises in Alberta. This could be prompted, for example, by a decision to build a new bitumen upgrader in the Edmonton area.

Alternatively, major markets for hydrogen were identified in California, the U.S. midwest and farther east. In a report prepared by Law, Sigurdson & Associates, it was noted that this market would result mostly from the need to refine gasoline and diesel fuel to higher standards. Also, it is believed that in the future it will be cheaper to produce hydrogen in Alberta and transport it to markets in the U.S. than to use on-site plants in the U.S. to make hydrogen from natural gas. It was estimated that refineries in California, and others that are along the pipeline route from Alberta to California, will require an additional 500 million scfd of hydrogen by the year 2000. Movement of hydrogen/natural gas blends, in which the hydrogen component is approximately 10 to 20 per cent, does not appear to be attractive at present, however. This is largely owing to the lack of economical hydrogen-recovery systems and by concerns about the effects of hydrogen on the components of compressors. If the blends contain low concentrations of hydrogen, however, the mixtures might be used as fuels in niche markets.

On the other hand, the magnitude of the California market for additional hydrogen is so large, it may justify construction of a new, dedicated hydrogen pipeline. If this pipeline were to go ahead, it would be needed before 1996. By then, California refiners will be needing increased amounts of hydrogen. Thus, it was recommended that this option be explored immediately before the opportunity is lost.

Forming the basis of these investigations were data generated by DB Robinson Research Ltd. on the thermodynamic and combustion characteristics of various blends of hydrogen and natural gas, up to a maximum of 20 per cent hydrogen. Much of this information resulted from using the Peng-Robinson equations of state, and data were produced for compressibility, density, heat capacity, entropy, thermal conductivity, viscosity, diffusivity and enthalpy. Also, information on the following combustion characteristics was reported: gas engine and gas turbine performance, heat of combustion, flame velocity, explosive limits, auto-ignition temperature, heating equipment performance, and the generation of CO_2, CO and NO_x in turbines, engines and flames.

SKM Consulting Ltd. investigated the sizing of pipelines and compressors that would be required for hydrogen/natural gas mixtures, as well as the specifications of materials that would be necessary and the risks associated with this type of service.

It was concluded that more information is needed about the mechanism of hydrogen embrittlement in pipeline systems, and there is a need for predictive hazard analysis techniques for all mechanical systems that would be affected by hydrogen/natural gas mixtures. SKM also studied the concepts and costs relating to the purification of by-product hydrogen and storage in salt caverns. The investigators found that an economic method for recovering hydrogen from dilute blends of hydrogen/natural gas has not been developed at present.

In the study of fuel cells carried out by Hydro-Québec, it was assumed that a steam-methane reformer would be integrated with a phosphoric acid fuel cell. The latter was chosen because it is the only type of fuel cell system that has a demonstrated capability to use hydrogen/natural gas mixtures on a commercial scale. It was found that this concept is not economically attractive at present.

Overall, the project findings pointed to the need for more technical information about the design and operation of pipelines that would transport blends of hydrogen and natural gas, although the economic aspects of such movement are not attractive at present. In contrast, the use of by-product hydrogen within Alberta is quite attractive. Furthermore, high-purity hydrogen can be delivered to the California market at prices that are competitive with making hydrogen there now from steam-methane reforming of natural gas.

Publications

Colt Engineering Corporation. 1991. Hydrogen Transportation Using Present Pipeline Network Study. Volume III. Collection, Distribution and Use in Alberta. Prepared for Hydrogen Industry Council.

Transportation Using Present Pipeline Network Study. 1991. Volume I. Thermodynamic and Combustion Data. Prepared for Hydrogen Industry Council.

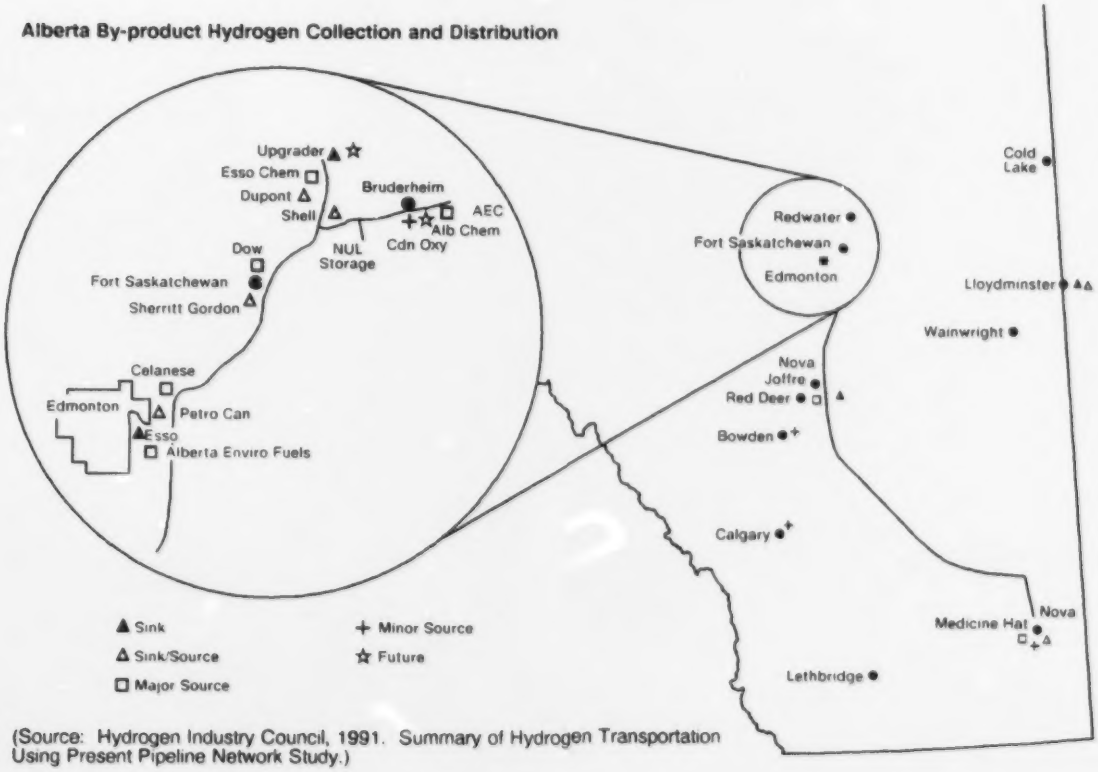
Hydrogen Industry Council. 1991. Summary of Hydrogen Transportation Using Present Pipeline Network Study.

Law, Sigurdson & Associates. 1991a. Hydrogen Transportation Using Present Pipeline Network Study. Volume IV. Collection, Distribution and Use Beyond Alberta. Prepared for Hydrogen Industry Council.

Law, Sigurdson & Associates. 1991b. Hydrogen Transportation Using Present Pipeline Network Study. Volume V. Collection, Distribution and Use Beyond Alberta. Appendices. Prepared for Hydrogen Industry Council.

SKM Consulting Ltd. 1991. Hydrogen Transportation Using Present Pipeline Network Study. Volume II. Pipeline and Compressors Purification, Storage and Generation. Hydrogen-Natural Gas Mixture for Fuel Cells. Prepared for Hydrogen Industry Council.

Alberta By-product Hydrogen Collection and Distribution



(Source: Hydrogen Industry Council, 1991. Summary of Hydrogen Transportation Using Present Pipeline Network Study.)

Hydrogen Use

Although oil refining and synthetic crude oil production rank among the largest uses of hydrogen in Alberta, even greater quantities are used by fertilizer and methanol producers. Hydrogen is also used in manufacturing chemicals and pharmaceuticals, and is important in steel making and metallurgy. Thus, the Alberta Hydrogen Research Program encompasses all possible uses for hydrogen in the province, with equal emphasis on improving the efficiency of current hydrogen-consuming processes and finding new uses for hydrogen in Alberta.

Two research projects related to this aspect of the program were under way during 1991/92.

Optimization of Hydrogen Utilization in Heavy Hydrocarbon Processing[1]

ALBERTA CHAMBER OF RESOURCES, EDMONTON

One of the original premises of the Alberta Hydrogen Research Program was that Canada will need more crude oil from non-conventional sources, such as the oil sands, as reserves of conventional light and medium crude oils decline. In turn, this will create a greater demand for hydrogen, which is used to upgrade synthetic crude oils to refinery-acceptable feedstocks. To keep the costs of upgrading as low as possible, it is necessary to make the current methods of using hydrogen more cost-efficient, or find new or improved ways of using hydrogen.

Thus, in a project coordinated by the Oil Sands Task Force of the Alberta Chamber of Resources, and carried out by RTM Engineering Ltd., the objective was to review and then select new or emerging technologies that have demonstrated some promise for improving the use of hydrogen in upgraders and refineries. Also, the project updated the hydrogen inventory study completed in 1988.

It was assumed that 15 800 m^3/day of Cold Lake bitumen would be processed in a new upgrader located near Fort Saskatchewan and Redwater, with startup in the year 2000. This upgrader was treated as either a standalone unit or in conjunction with a refinery that produced motor gasoline, jet fuel and diesel fuel. The actual refinery products were those projected to be in demand in 2000.

It was also assumed that both configurations of the upgrader would be expected to meet certain projected environmental standards. This would have a bearing on the types of pollution control systems used by either configuration. Also, environmental standards that are more stringent than those applicable today would influence the characteristics of petroleum products and their relative position in the marketplace.

It was projected that many changes would occur in both the quality of, and demand for, refined products. Higher hydrogen content and lower sulphur and aromatic content were likely. Gasoline consumption would decline, while use of jet fuel and diesel fuel would rise, making it difficult for current refineries to meet the projected demand. Changes in quality requirements would compound this problem and force significant investment at most existing refineries.

These changes in both the demand for and composition of petroleum products are expected to lead to a hydrogen shortage at current refineries unless on-site, steam-methane reforming of natural gas occurs.

Consequently, a combination upgrader/refinery was considered to be the more promising option. It would:

- eliminate duplication of hydrogen processing units for naphtha, jet fuel, diesel fuel and gas oil;
- provide common hydrogen production and recovery facilities;
- avoid the necessity for crude distillation occurring at a separate refinery;
- allow residual products from refining to be converted in the upgrader; and
- provide other synergistic benefits for upgrading and refining.

Publications

McCann, T.J. 1991a. Optimum Hydrogen Use in Upgrading and Refining. Phase I Report. Prepared for Oil Sand Task Force, Alberta Chamber of Resources. RTM Engineering Ltd.

McCann, T.J. 1991b. Optimum Hydrogen Use in Upgrading and Refining. Prepared for Oil Sand Task Force, Alberta Chamber of Resources. RTM Engineering Ltd.

[1]Funded jointly by Alberta Chamber of Resources and A/CERRF.

Hydrogen Utilization in Bitumen Upgrading[1]

ALBERTA CHAMBER OF RESOURCES, EDMONTON

Following completion of the Optimization of Hydrogen Utilization in Heavy Hydrocarbon Processing project, a new project was initiated in 1991. Its objective is to gain a better understanding of how hydrogen reacts with bitumen when the latter is upgraded and refined to produce transportation fuels. The long-term objective is to increase product quality without increasing hydrogen consumption.

The study is employing deuterium labelling and nuclear magnetic resonance and mass spectroscopy to track the fate of the labelled species during primary and secondary bitumen upgrading. This should help provide information about the mechanisms involved when hydrogen reacts with particular carbon atoms and to what extent hydrogen wastage can be reduced.

The investigation is using the specialized equipment and services available at the Energy Research Laboratories of the Canada Centre for Mineral and Energy Technology (CANMET) near Ottawa. Direction is being provided by the Oil Sands Task Force of the Alberta Chamber of Resources.

Thus far, the products of low- and high-severity hydrocracking have been analysed. A difference in H_2 exchange between low- and high-severity conditions was detected. Also, analysis of the naphtha products of low-severity hydrocracking showed an increased incorporation of deuterium into the methyl groups that are α to aromatic rings, methylene groups that are either α to olefinic CH groups or cyclo-paraffinic groups, and both olefinic CH groups and CH species of alkyl-substituted cyclo-paraffin groups.

[1]Funding was provided by the Alberta Chamber of Resources, CANMET and the Alberta Department of Energy.

Publications

Dettman, H.D. and D.J. Patmore. 1992. Hydrogen Utilization in Bitumen Upgrading: An Experimental Study of the Fate of Process Hydrogen. Second Quarter Report. CANMET.

Dettman, H.D. and D.J. Patmore. 1991. Hydrogen Utilization in Bitumen Upgrading: An Experimental Study of the Fate of Process Hydrogen. First Quarter Report. CANMET.

Other

University of Calgary Hydrogen Industry Chair

THE UNIVERSITY OF CALGARY, CALGARY

A position at The University of Calgary is being established with a grant from A/CERRF, some industry participants[1] and the Natural Sciences and Engineering Research Council of Canada. The objective is to conduct hydrogen research that will be of direct benefit to the industry partners.

At year-end, discussions were under way regarding selection of the chairholder.

[1]Research funds contributed by A/CERRF in 1991 are being matched by the following group of industry participants: Shell Canada Limited, Esso Resources Canada Limited, Husky Oil, Gulf Canada Resources Ltd, AMOCO Canada Petroleum Company, TransAlta Utilities Corporation and Alberta Oil Sands Technology and Research Authority. Also, substantial funding is being provided by the Natural Sciences and Engineering Research Council of Canada. The University of Calgary is contributing the salary of one professor.

Hydrogen Research Program Workshop

INFO-TECH CONFERENCE MANAGEMENT, CALGARY

On November 7, 1991, the Alberta Department of Energy sponsored a one-day workshop on hydrogen research projects funded by the department and industry. This was the fourth such workshop. Eleven presentations were made to an audience of 49 attendees. Based on an enthusiastic response by those who attended, another similar conference is planned for 1992.

Titles of the conference presentations are listed below.

Hydrogen Production

A Claus Plant Modification for Hydrogen Production

Cold Plasma Decomposition of Hydrogen Sulphide

The CO-Catalyzed Conversion of H_2S to H_2 and Sulphur

Hydrogen Sulphide Electrolysis in Liquids

Chemically Modified Electrodes for Hydrogen Sulphide Electrolysis

High-Temperature Electrolysis of Aqueous Hydrogen Sulphide for the Production of Hydrogen and Molten Sulphur

Hydrogen Separation

Hydrogen Separation and Purification

High-Temperature Separation of Hydrogen Using Ceramic Membranes

Hydrogen Transportation

Hydrogen Transportation Using Present Pipelines Network

Hydrogen Use

Optimum Hydrogen Utilization in Upgrading and Refining

By-product Hydrogen to Electricity via Fuel Cells

Appendices

All research projects supported by the Alberta Hydrogen Research Program are listed in Appendix A, while information about the funding provided by A/CERRF and the Department of Energy is found in Appendix B.

Appendix A

Complete Listing of Projects Supported by the Alberta Hydrogen Research Program

Project	Researcher	Status
Hydrogen Production		
Examination of the Partial Oxidation of Methane for the Production of Hydrogen and/or Synthesis Gas	G.A. Karim, The University of Calgary, Calgary	Completed in 1989/90
A Claus Plant Modification for Hydrogen Production	Alberta Sulphur Research Ltd., Calgary	Continuing
Conversion of Hydrogen Sulphide to Hydrogen and Organosulphur Compounds by Metal Catalysts	Alberta Sulphur Research Ltd., Calgary	Completed in 1990/91
Production of Hydrogen and Sulphur from Hydrogen Sulphide	L.G. Hepler, University of Alberta, Edmonton and J. Donini, CANMET, Devon	Completed in 1990/91
High-Temperature Electrolysis of Aqueous Hydrogen Sulphide for the Production of Hydrogen and Molten Sulphur	Alberta Research Council, Edmonton	Completed in 1990/91
Chemically Modified Electrodes for Hydrogen Sulphide Electrolysis	Alberta Research Council, Edmonton	Completed in 1991/92
The Carbon Monoxide Catalyzed Conversion of Hydrogen Sulphide to Hydrogen and Sulphur	O.P. Strausz, University of Alberta, Edmonton	Continuing
Cold Plasma Decomposition of Hydrogen Sulphide	Shell Canada Limited, Calgary and Atomic Energy of Canada Limited, Chalk River	Continuing
Microwave-Initiated Decomposition of Hydrogen Sulphide	O.P. Strausz, University of Alberta, Edmonton	Continuing

Hydrogen Separation

Hydrogen Separation and Purification	Alberta Research Council, Edmonton	Completed in 1990/91
High-Temperature Separation of Hydrogen Using Novel Ceramic Membranes	Alberta Research Council, Edmonton	Continuing

Infrastructure, Storage and Transportation

Hydrogen Producer/ Consumer Network	Alberta Research Council, Devon	Completed in 1990/91
Salt Cavern Storage of Hydrogen	RTM Engineering Ltd., Calgary	Completed in 1990/91
Hydrogen Transportation Using Present Pipeline Networks	Hydrogen Industry Council, Calgary	Completed in 1991/92

Hydrogen Use

Optimization of Hydrogen Utilization in Heavy Hydrocarbon Processing	Alberta Chamber of Resources, Edmonton	Completed in 1991/92
Refinery By-product Hydrogen Use for Electricity Generation in Fuel Cells	TransAlta Utilities Corporation, Calgary	Completed in 1990/91
Hydrogen Utilization in Bitumen Upgrading	Alberta Chamber of Resources, Edmonton	Continuing

Other

Hydrogen Technology Program Planning and Implementation Assistance	Harold V. Page, Edmonton	Completed in 1990/91
University of Calgary Hydrogen Industry Chair	The University of Calgary, Calgary	Continuing
Hydrogen Research Program Workshop	Info-Tech Conference Management	Completed in 1991/92

Appendix B

Alberta Hydrogen Research Program
Summary of Expenditures ($)

PROJECT	1987/88	1988/89	1989/90	1990/91	1991/92	Projected 1992/93	TOTAL
Hydrogen Production							
A Claus Plant Modification for Hydrogen Production	-	-	68 490	143 847	145 836	164 000	522 173
Production of Hydrogen and Sulphur from Hydrogen Sulphide	-	-	76 235	70 665	-	-	146 900
Conversion of H_2S to H_2 and Organosulphur Compounds by Metal Catalysts	-	-	12 806	17 194	-	-	30 000
Examination of the Partial Oxidation of Methane for Production of Hydrogen and/or Synthesis Gas	-	-	58 391	-	-	-	58 391
High-Temperature Electrolysis of Aqueous Hydrogen Sulphide	-	-	-	60 000	-	-	60 000
The CO-Catalyzed Conversion of H_2S to H_2 & Sulphur	-	-	-	81 640	13 800	24 500	119 940
Cold Plasma Decomposition of H_2S	-	-	-	194 627	194 819	37 749	427 195
Chemically Modified Electrodes for Hydrogen Sulphide Electrolysis	-	-	-	25 393	26 488	-	51 881
Microwave-Initiated Catalytic Decomposition of Hydrogen Sulphide	-	-	-	-	60 794	57 878	118 672
Electrolysis of H_2S in Alkaline Solution	-	-	-	-	-	42 569	42 569
Hydrogen Decomposition Using Modified Aluminas	-	-	-	-	-	38 665	38 665
Subtotal Hydrogen Production	**0**	**0**	**215 922**	**593 366**	**441 737**	**365 361**	**1 616 386**
Hydrogen Storage and Transportation							
Salt Cavern Storage of Hydrogen	-	-	43 071	-	-	-	43 071
Hydrogen Producer/Consumer Network	-	-	39 492	-	-	-	39 492
Hydrogen Transportation, Using Present Pipeline Networks	-	-	-	56 324	77 525	-	133 849
Subtotal Hydrogen Storage and Transportation	**0**	**0**	**82 563**	**56 324**	**77 525**	**0**	**216 412**

PROJECT	1987/88	1988/89	1989/90	1990/91	1991/92	Projected 1992/93	TOTAL
Hydrogen Producers/Consumers							
Hydrogen Separation and Purification	-	-	167 342	116 454	-	-	283 796
High-Temperature Separation of H_2 Using Novel Ceramic Membranes	-	-	-	58 511	65 185	81 830	205 526
High-Efficiency Hydrogen Separation and Purification	-	-	-	-	-	93 250	93 250
Subtotal Hydrogen Producers/Consumers	**0**	**0**	**167 342**	**174 965**	**65 185**	**175 080**	**582 572**
Hydrogen Utilization							
Optimization of H_2 Utilization in Heavy Hydrocarbon Processing	-	-	-	93 246	-	-	93 246
Refinery By-product Hydrogen Use for Electrical Generation in Fuel Cells	-	-	-	15 075	-	-	15 075
Hydrogen Utilization in Bitumen Upgrading: A Study on Process Hydrogen	-	-	-	-	43 000	-	43 000
Study on Upgrader Optimization	-	-	-	-	-	35 000	35 000
Multistage Hydrotreating of Athabasca Bitumen and Cracked Products	-	-	-	-	-	130 000	130 000
Wide-Cut Cold Lake Blend Distillate Hydrotreating Study	-	-	-	-	-	43 500	43 500
Hydrogen Peroxide by Direct Oxidation of Hydrogen	-	-	-	-	-	40 000	40 000
Subtotal Hydrogen Utilization	**0**	**0**	**0**	**108 321**	**43 000**	**248 500**	**399 821**
Hydrogen Special Projects							
Hydrogen Technology Inventory Study for Alberta	47 874	-	-	-	-	-	47 874
Hydrogen Technology Program Planning and Implementation Assistance	-	5 805	-	-	-	-	5 805
University of Calgary Hydrogen Industry Chair	-	-	-	375 000*	-	-	375 000
Hydrogen Workshop 1991	-	-	-	-	4 950	-	4 950
Hydrogen Workshop 1992	-	-	-	-	-	6 000	6 000
Subtotal Hydrogen Special Projects	**47 874**	**5 805**	**0**	**375 000***	**4 950**	**6 000**	**439 629**
TOTAL Alberta Hydrogen Research Program	**47 874**	**5 805**	**465 827**	**1 307 976***	**632 397**	**794 941**	**3 254 820**

* Advanced transfer of $375 000 to The University of Calgary Hydrogen Industry Chair

Additional copies of this review are available from:

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